Low-Temperature Architected Cementation Agents (LAMINAE)

Gaurav N. Sant, UCLA

**Team Members: Dante Simonetti, Mathieu Bauchy, Samanvaya Srivastava, Juan Carlos Vega-Vila, Ross Arnold (UCLA)
Aditya Kumar (MST)
Bu Wang (UW-M)
Yann Le Pape, Elena Tajuelo Rodriguez (ORNL)**

Project Goal

A low-temperature, low-energy, low-CO_2 pathway to microstructure-controlled zeolitic cementation agents
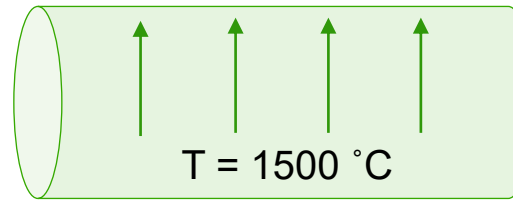
Total project cost:	$1.9M
Current Q / Total Project Qs	9 / 12

The Concept: Low-Temperature Architected Cementation Agents

Current Technology to Produce OPC

Calcium-aluminate
Calcium-silicate
Calcium-ferrite
Gypsum

Thermal activation of carbonate-silicate-aluminate
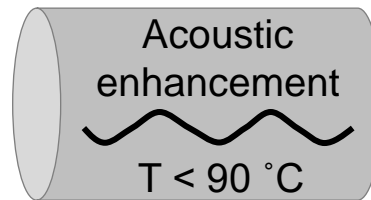
T = 1500 °C

Ordinary Portland Cement (OPC)

- 0.90 kg CO_2 (kg OPC)$^{-1}$
- 6000 J g^{-1}

LAMINAE

Geological precursor

Acoustically-enhanced precursor dissolution

Acoustic enhancement

T < 90 °C

Si, Al, Ca, Na

Ultrafast silicate hydrate synthesis

Rapid heating

T < 200 °C
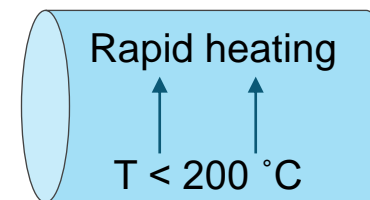
Cementitious zeolites
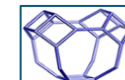
- 0.35 kg CO_2 (kg zeolite)$^{-1}$
- < 4000 J g^{-1}

Low Energy: Reduced by 30% vs. OPC	**Low CO_2:** Reduced by 60% vs. OPC	**Controlled Microstructure:** Non-passive hydration improves ductility, durability

The Team

UCLA Samueli School of Engineering

Gaurav N. Sant
Project Lead

Dante Simonetti
Process engineering, reactor design, chemical kinetics

Mathieu Bauchy, Samanvaya Srivastava, Juan Carlos Vega-Vila, Ross Arnold

Process engineering, reactor design, chemical kinetics, zeolite synthesis and characterization, molecular dynamics simulations
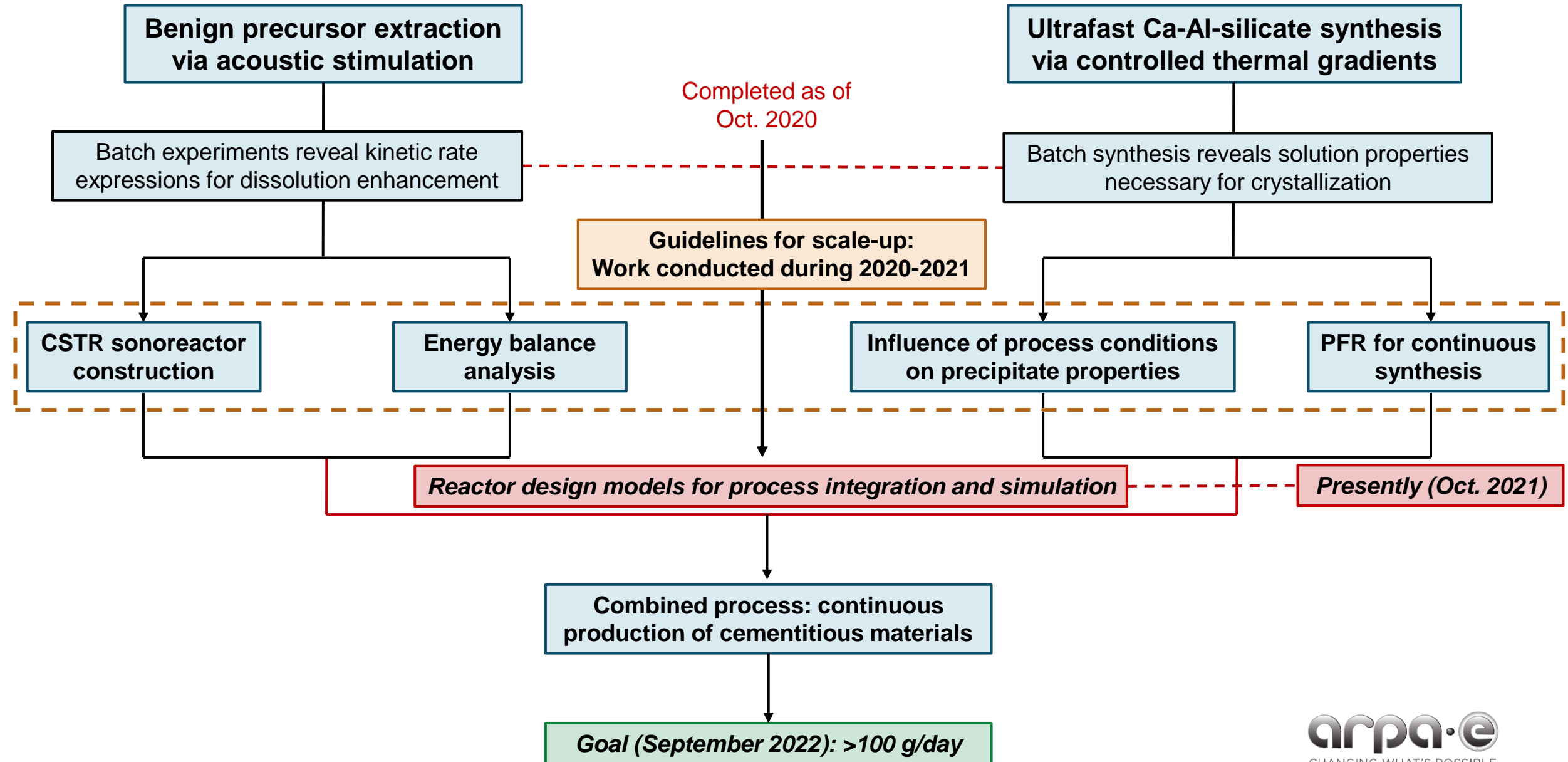
MISSOURI S&T University of Science & Technology

Aditya Kumar
Reaction kinetics

WISCONSIN UNIVERSITY OF WISCONSIN–MADISON

Bu Wang
Material adhesion and cohesion

OAK RIDGE National Laboratory

Yann Le Pape,
Elena Tajuelo Rodriguez
Characterization

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# Project Objective: Continuous Production of Cementitious Materials

Results – Reaction Chemistry

▸ The identity and structural properties of metal cations that participate in zeolite precipitation, in the absence of organic structure-directing agents, determine the crystallized zeolite topology

– Calcium facilitates tobermorite crystallization, whereas sodium and potassium facilitate phillipsite crystallization

▸ Phillipsite crystallization and precipitation depends on the silicon concentration on synthesis gels

– Phillipsite crystallized at varied silicon concentrations (0.2 – 3 M)

– Similar silicon concentration range for phillipsite crystallization in batch and flow reactor
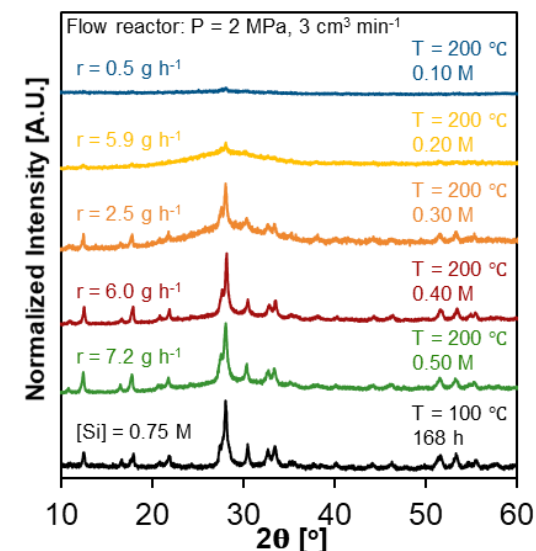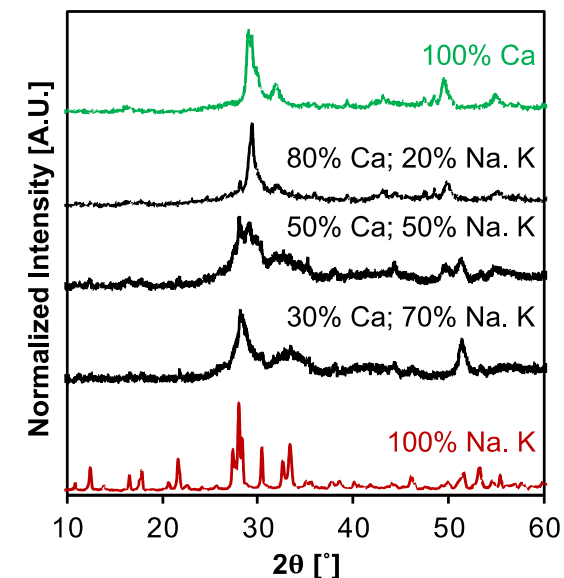
Results – Process Kinetics and Reactor Design

▶ Batch and flow reactor Si dissolution kinetics can be modeled using zero-order dissolution and first-order coprecipitation kinetics:

- $r_{dissolution} = k_1; r_{precipitation} = k_2[\text{Si}]$

- Model allows for evaluation of effect of temperature, pH, residence time on continuous stirred tank sonoreactor

● Zeolite rate of production from batch syntheses is directly proportional to Si concentration, and inhibited by K concentration

- $r_{precipitation} = k_{eff} \dfrac{[\text{Si}]^a}{[\text{K}]^d}$

- Model serves as basis for design of plug flow zeolite precipitation reactor
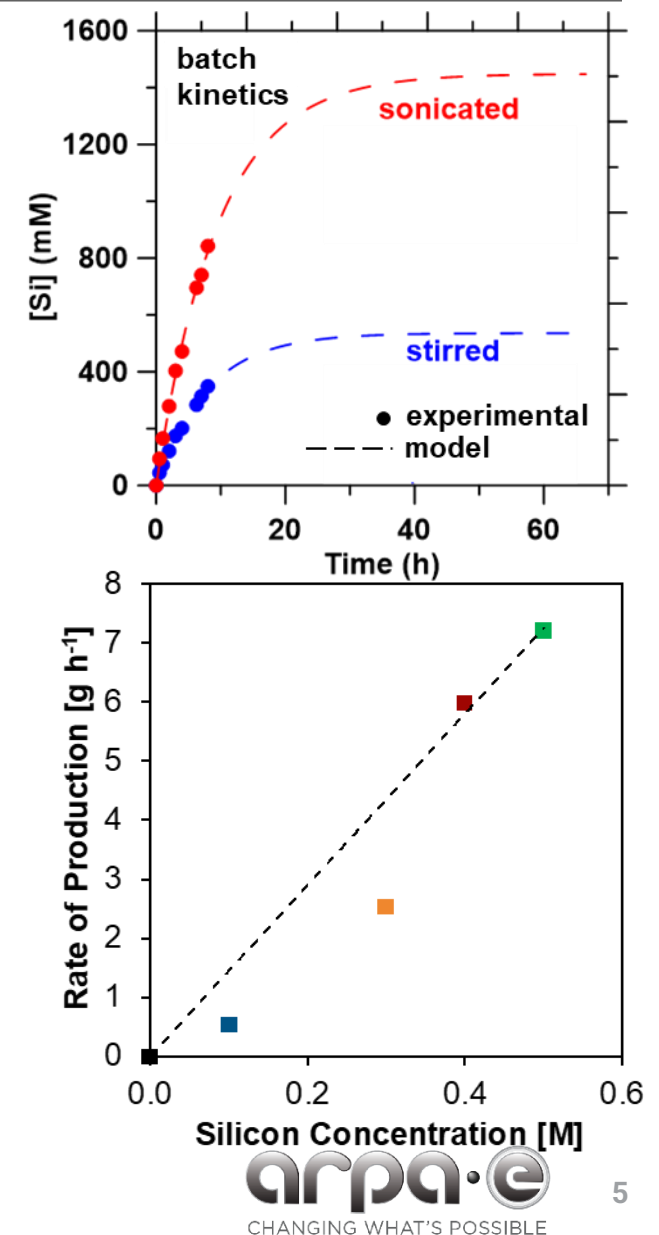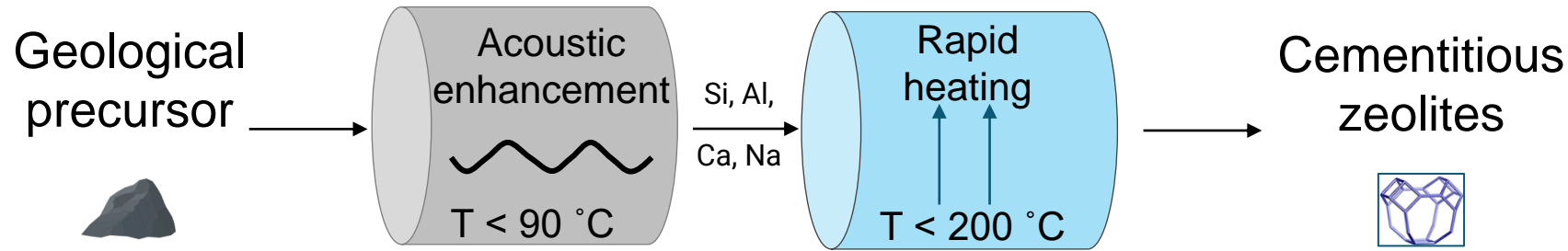
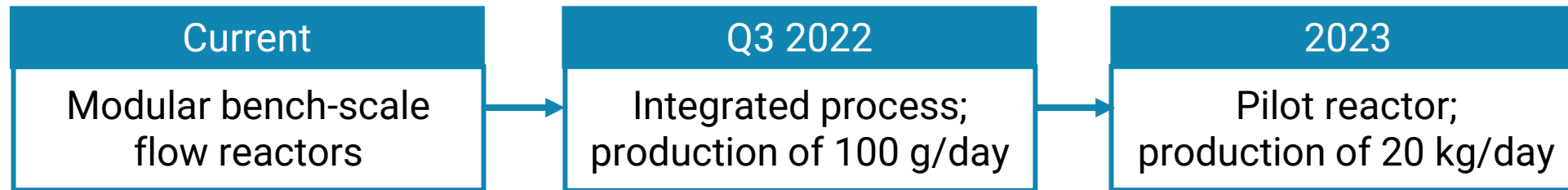# Challenges, Risks and Potential Partnerships

- **Challenges** associated with continuous production of cementitious agents

  - Acoustic Dissolution-Hydrothermal Synthesis mismatch
    - **Challenge:** Effluent of the sonoreactor contains <1 mM Si, whereas synthesis reactor requires an influent at 12-200 mM Si
    - **Solution:** Boost the pH of sonoreactor effluent; Recycle stream containing crystallization "seeds"
  - Reactor Design and Optimization
    - **Challenge:** Will heat-mass transfer impact CSTR-PFR at larger scale?
    - **Solution:** CFD modeling to identify gradients; Research falling film and fluidized bed reactors
  - Resistance to adoption of new technology
    - **Challenge:** OPC and catalyst/sorbent production have remained unchanged for decades
    - **Solution:** Bench-scale demonstration and product testing are critical to increasing confidence of potential adopters

- We are interested in **partnerships** with others offering expertise in high-pressure thermal analysis (including differential scanning calorimetry), and high pressure and temperature kinetic modeling of dissolution and precipitation processes

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Technology-to-Market

‣ The LAMINAE process will be licensed as a commercial technology:

Geological precursor → [Acoustic enhancement, T < 90 °C] → Si, Al, Ca, Na → [Rapid heating, T < 200 °C] → Cementitious zeolites

‣ Process is estimated at two years from pilot-scale demonstration

Current	Q3 2022	2023
Modular bench-scale flow reactors	Integrated process; production of 100 g/day	Pilot reactor; production of 20 kg/day

‣ Process will be marketed for both alternative cement production and catalyst and sorbent production applications

‣ Progress can be accelerated through collaboration with partners with targeted expertise, such as industrial-scale reactor design

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Summary

Low Energy	Low CO2	Controlled Microstructure
Reduced by 30% vs. OPC	Reduced by 60% vs. OPC	Non-passive hydration improves ductility, durability
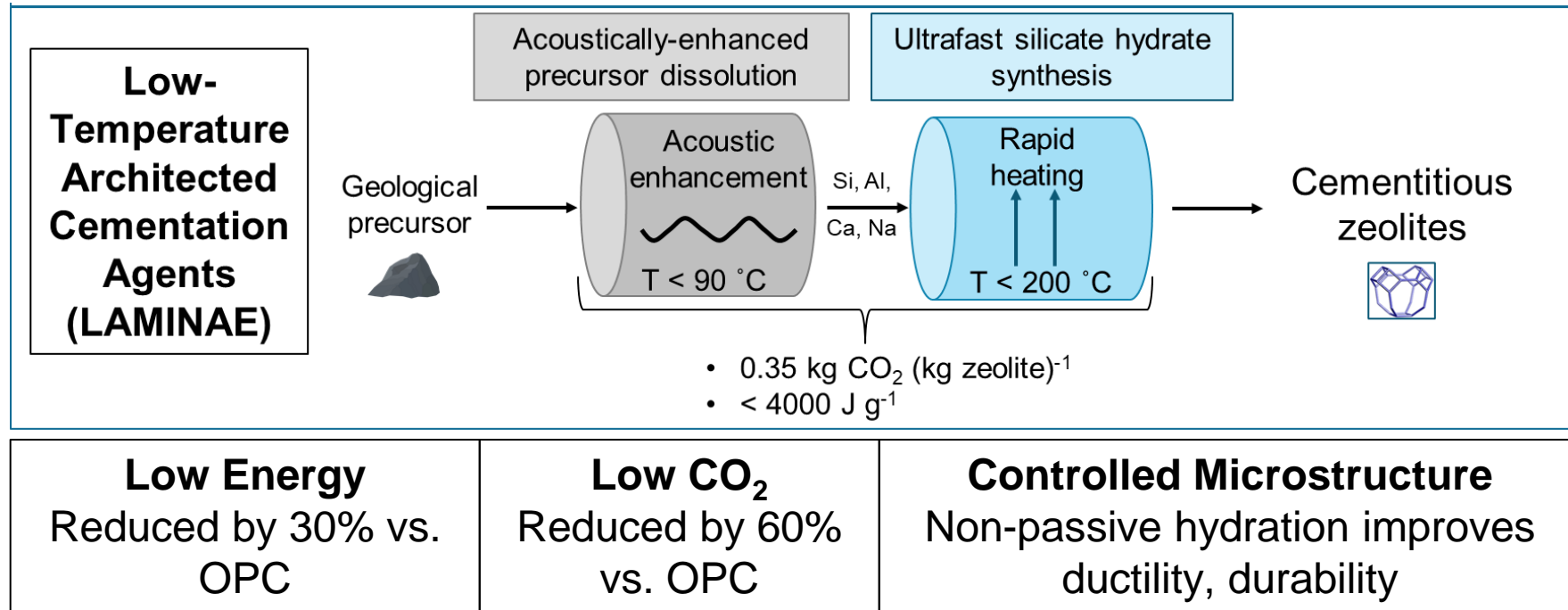
▸ Currently building bench-scale flow reactors using design equations derived from fundamental reaction kinetics measurements

▸ Final target of current project is an integrated bench-scale reactor, producing 100 g/day of cementitious zeolites by Q3 2022

https://arpa-e.energy.gov